A Closed Loop Process for the End-of-Life Electric Vehicle Li-ion Batteries: Phase II

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Relevance and Project Objectives

- During the course of the Phase I USABC program which focused on using recycled batteries to produce NMC111, the team has observed that the xEV battery industry is moving to higher nickel NMCs.

- Building on the successful Phase I program, the overall objective of the Phase II program is to demonstrate the recovery of NMC622 cathode materials from recycled lithium ion batteries with mixed cathode and anode chemistry, and added complexity of adhesives, silicon and LTO that are anticipated materials in the future waste stream.

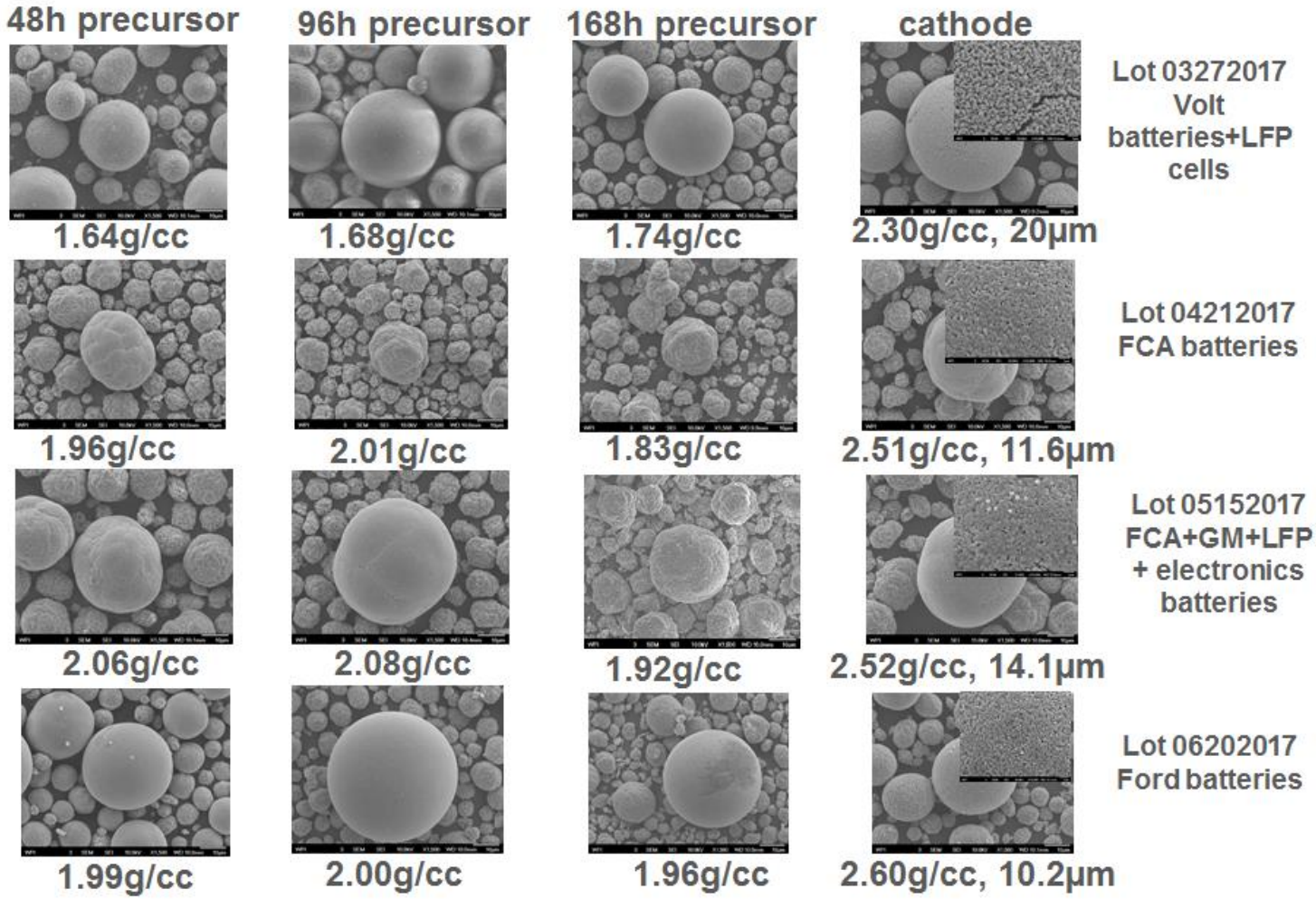
- The cost model developed for NMC111 will be updated based on the new chemistry process update and scale-up.

Phase I Summary: Overall Recycling Process

Advantages:

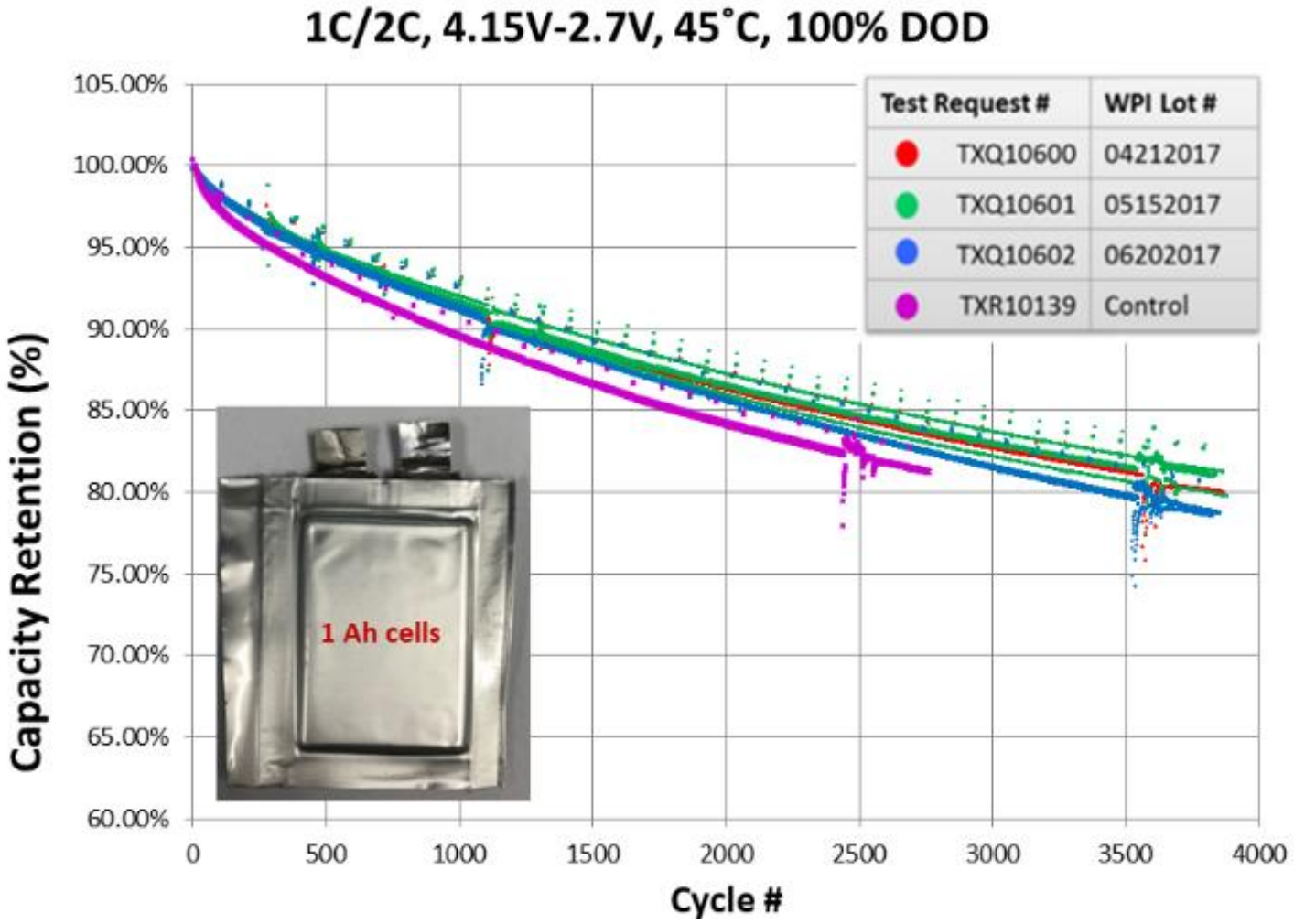
- Any lithium Ion battery
- Any size and shape
- No sorting
- Synthesize new $LiNi_xMn_yCo_zO_2$ directly
- Ratio of Ni, Mn and Co can be specially tailored to customer demands

Phase I Summary:
NMC111 from Different Recycling Streams



NMC111 powder has been synthesized from different recycling streams. The powder is used for 1Ah cells.

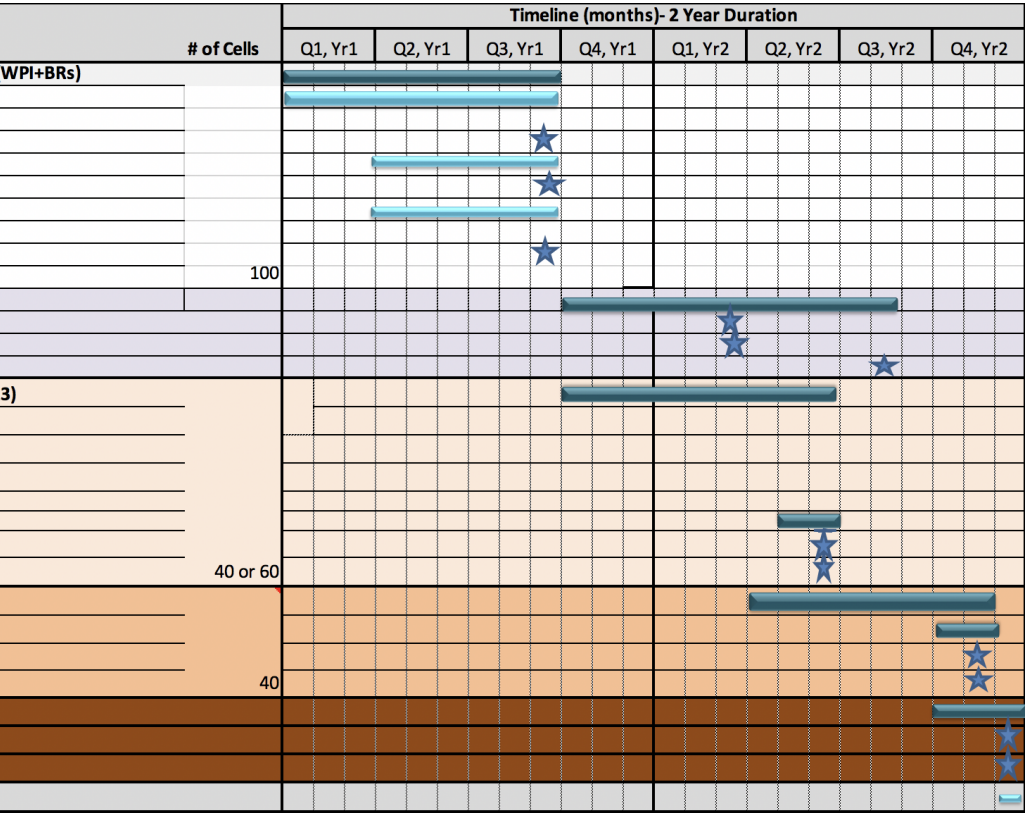
Phase I Summary: 1Ah Cell Performance



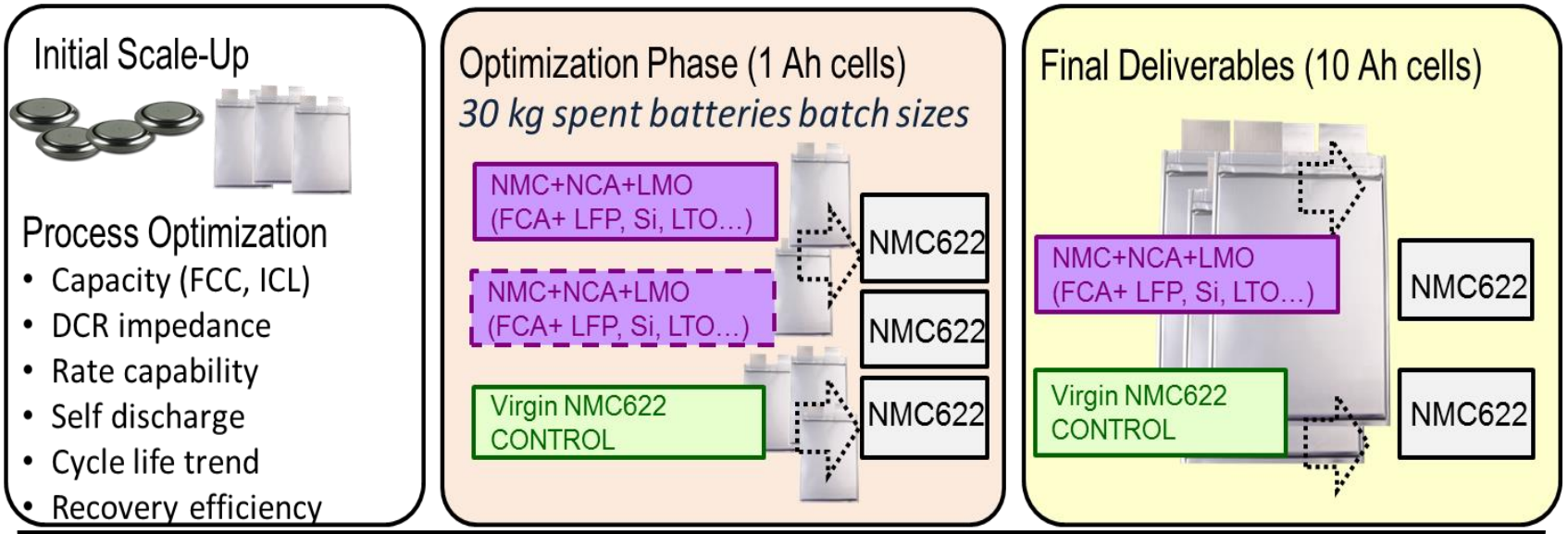
1Ah cells from different batches of WPI recovered NMC111 shows better performance than the commercial powder. These cells are tested at A123 Systems.

Phase II: Milestones

Tasks and Milestones	# of Cells
Task 1:Optimize the electrochemical properties of recycled NMC622 cathode materials (WPI+BRs)	
1.1 Optimize NMC622 synthesis parameters	
pH, material adding rate, temperature, etc are determined.	
Milestone 1.1: Recovered NMC622 shows similar properties to commercial NMC622	
1.2 Understand the impacts of new anode materials and possible adhesive	
Milestone 1.2:Determine the impacts of anode materials and adhesive	
1.3 Electrodes Development	
Cathode Evaluation: Coin Cell, Full-Cell	
Milestone 1.3: Half cell testing shows properties within 5% of virgin material	
100 coin cells fabricated to evaluate recycled NMC622	100
Task 2: Scale Process to 30 kg batch size (WPI+BRs)	
Milestone 2.1: Scaled process cathode powder within 5% of control	
Deliverable: WPI deliver 1 or 2 batches of 6kg recovered NMC622 to A123 for 1Ah cells	
Deliverable: WPI deliver 15kg recovered NMC622 to A123 for 10Ah cells	
Task 3: Produce 60 1Ah cells from recycled material (1 or 2 input stream+control) (A123)	
Cell Assembly and Formation	
Performance Testing	
Cycle life	
Calendar Testing	
Internal Screening	
Milestone 3.1 Recovered cathode cells within 5% of control cells	
Deliverable: 40 or 60 1Ah cells to USABC/A123	40 or 60
Task 4: Fabricate large format prismatic 10 Ah cells (1 input stream+control) (A123)	
Internal Screening	
Milestone 4.1 Recovered cathode cells within 5% of control	
Deliverable: 40 10Ah cells to USABC/A123	40
Task 5: Recycling process update (BRs)	
Milestone 5.1 Verify that the recycling process is economical feasible	
Deliverable: Updated recycling process and cost analysis	
Final Report (WPI)	



Phase II: Cell Fabrication and Test Plan

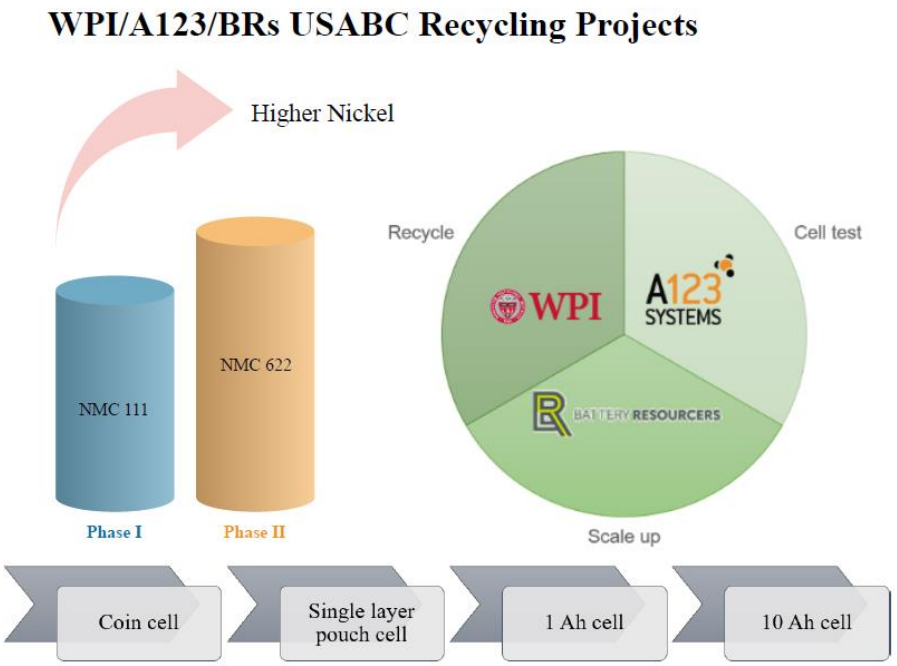


Build	Test Article	Total Made	USABC/ ANL	A123/ WPI	Kg powder per group
1	1 Ah NMC622/gr	20	12	8	6 (WPI)
1-control	1 Ah NMC622/gr	20	12	8	6 (comm.)
(2)*	(1 Ah NMC622/gr)*	(20)*	(12)*	(8)*	6 (WPI))*
3	10 Ah NMC622/gr	20	12	8	15
3-control	10 Ah NMC622/gr	20	12	8	15 (comm.)

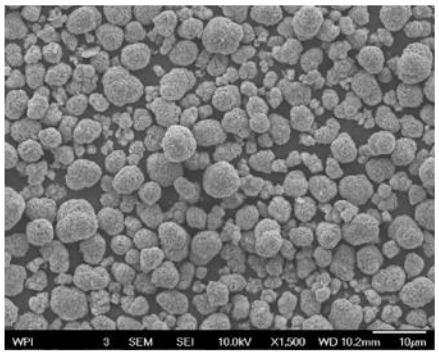
** 2nd 1 Ah build will only be executed if required. Depends upon 1st build results.*

Phase II: Approach/Strategy

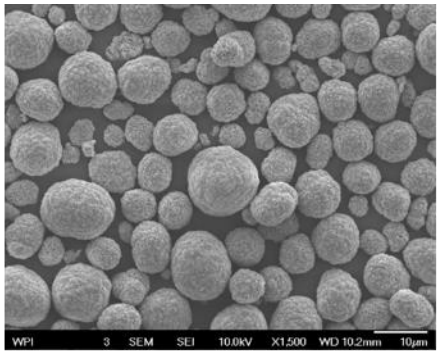
- Develop high nickel NMC from the spent EV batteries
- Understand the impacts of different anode materials and adhesives on the recycling process
- Develop scalable coating method for NMC622
- Recovered materials are tested in large pouch cells at A123 Systems
- The scale-up and cost model of the recycling process are developed at Battery Resourcers



Phase II: Technical Accomplishment and Progress



Day 1



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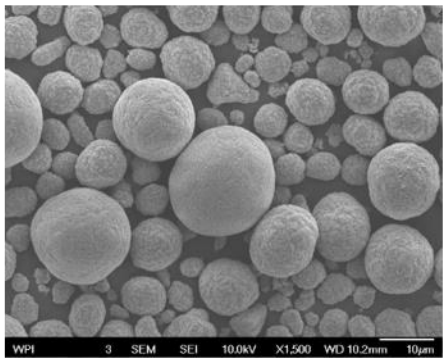

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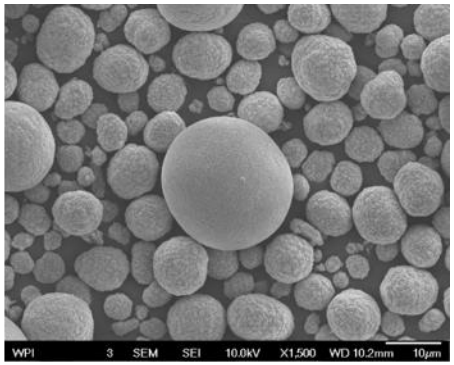

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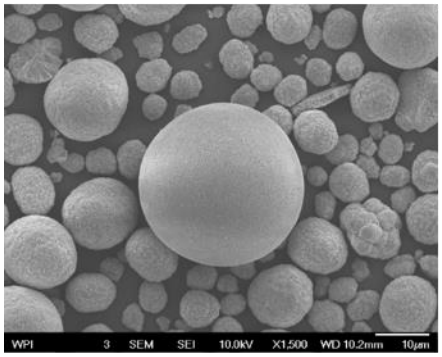

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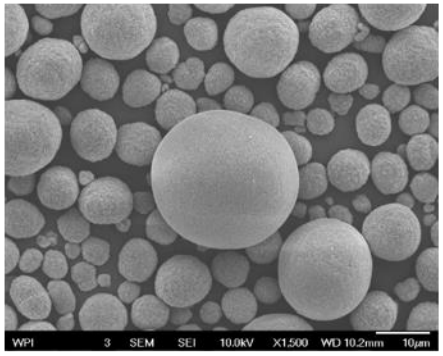

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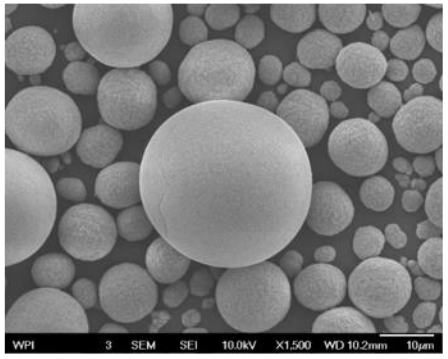

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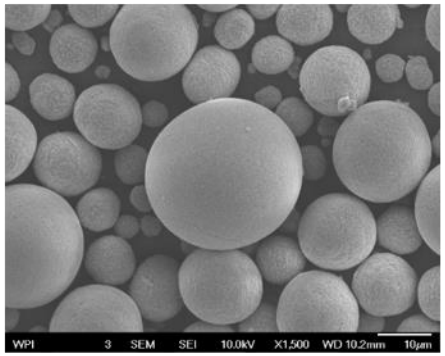


Main Reactor

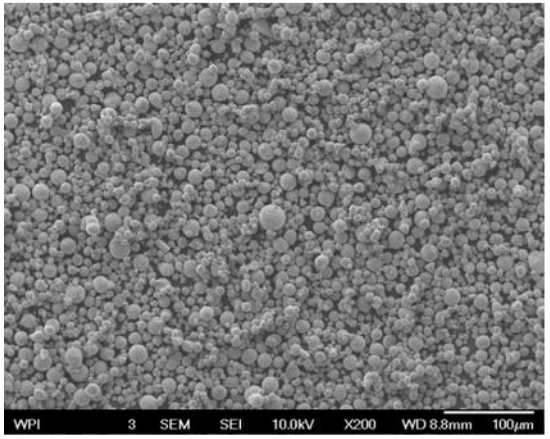
NMC622 precursor with good morphology has been synthesized with the recycled materials.

Phase II: Tap Density of NMC622 Precursor

	Weight/g	Tap Density/g·ml^{-1}
Day 1	311.5	1.85
Day 2	666.92	2.19
Day 3	752.68	2.35
Day 4	727.38	2.34
Day 5	636.62	2.33
Day 6	636.95	2.35
Day 7	175.56	2.34
Main	176.04	2.35
Aging	122.03	2.33

Phase II: Recycled NMC622 Cathode Powder

*200



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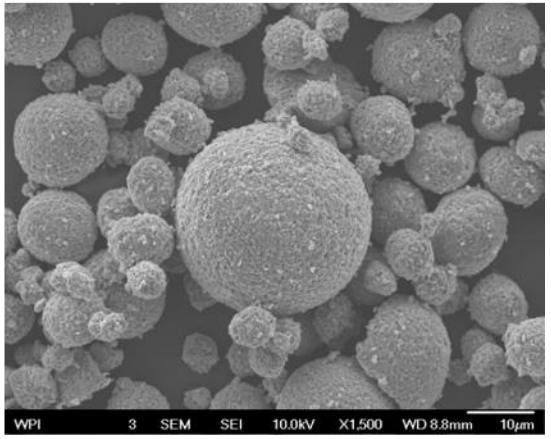

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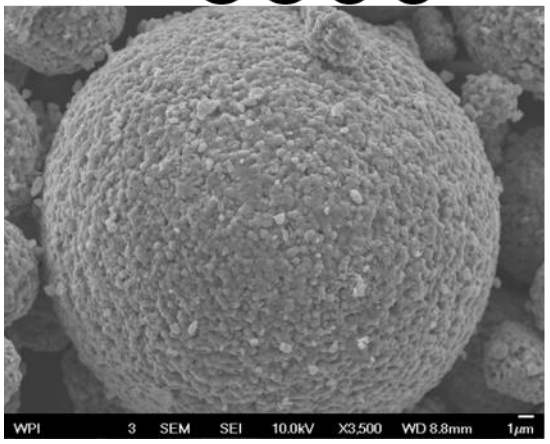

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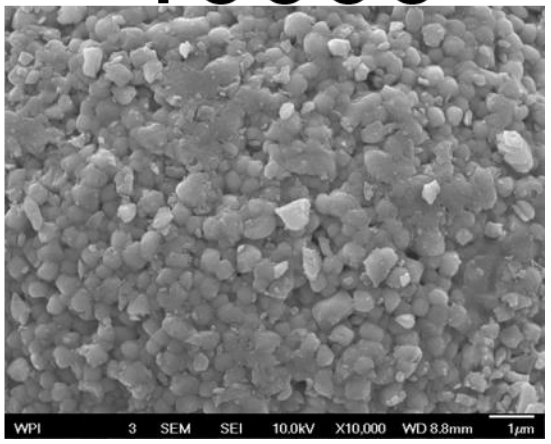
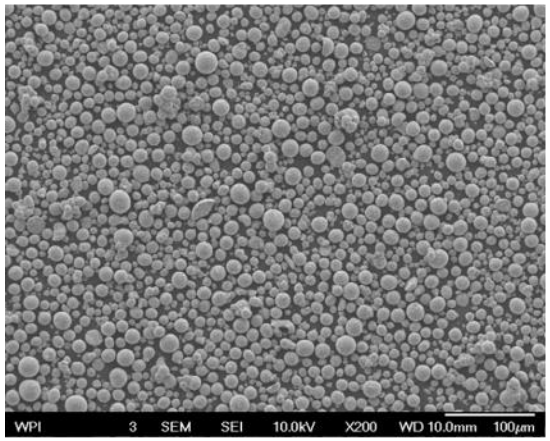


Lot# 10152018
2.72g/mL

Phase II: Coated NMC622 Cathode Powder 1

*200



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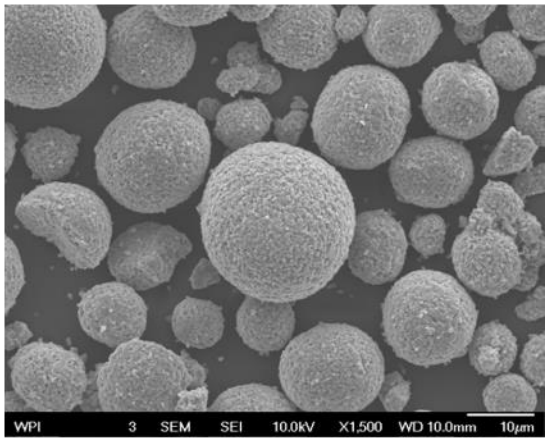

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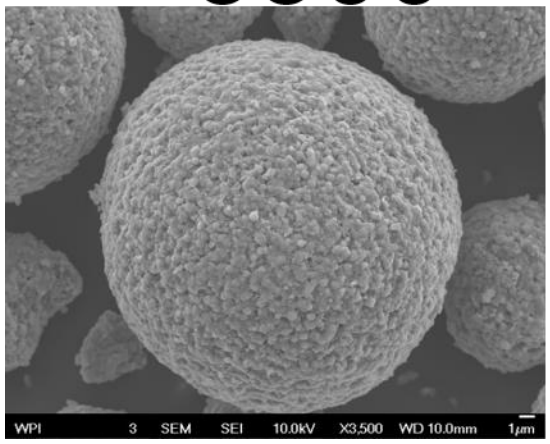

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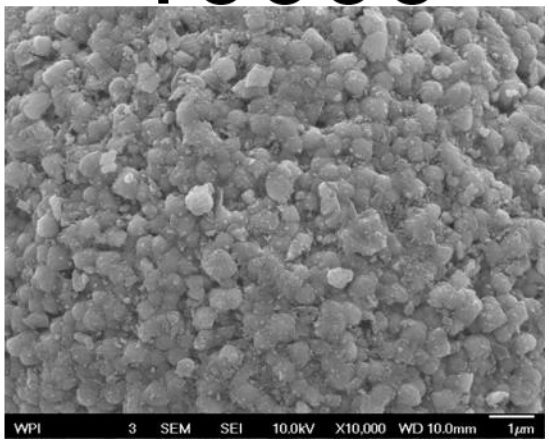

Lot# 10222018 -C1
2.78g/mL

Phase II: Coated NMC622 Cathode Powder 2

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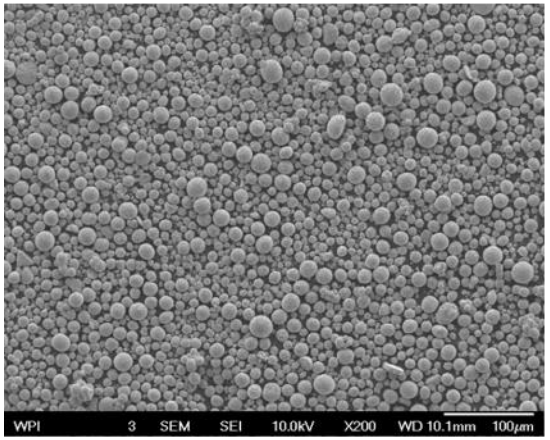

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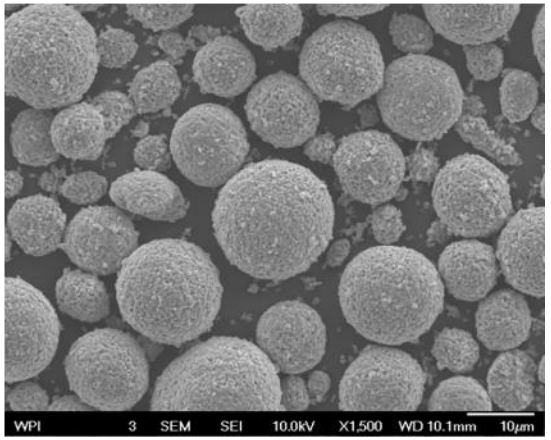

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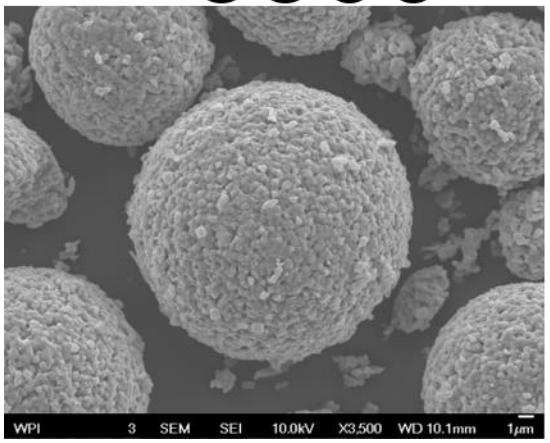

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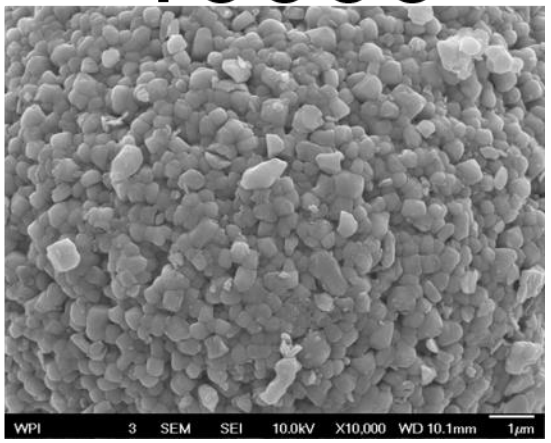


Lot# 10222018 -C2 2.82g/mL

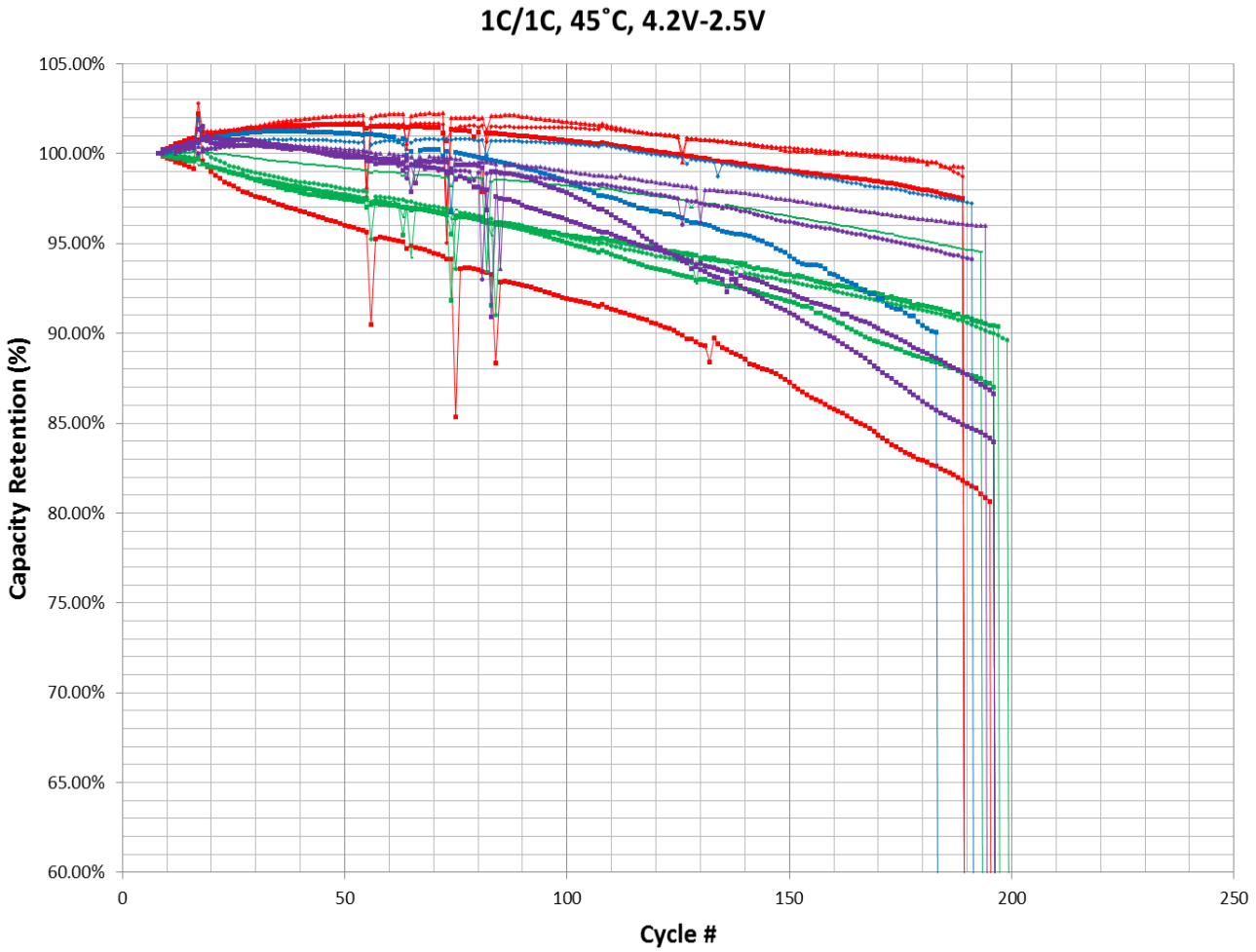
Phase II: A123 Test Results (Coin Cells)

Physical Properties					Electrode		Echem								
BET (m^2/g)	Tap Density (g/cm^3)	D10 (µm)	D50 (µm)	D90 (µm)	Load mg/cm2	Press Density (g/cc)	FCC (0.1C) (mAh/g)	FDC (0.1C) (mAh/g)	First Cycle Eff (%)	0.2C (mAh/g)	0.5C (mAh/g)	1C (mAh/g)	2C (mAh/g)	5C (mAh/g)	10C (mAh/g)
0.362	2.66	7.1	12.8	22.1	21.02	2.85	193.9	170.1	87.7	166.8	160.7	154.8	144.2	61.1	14.3
0.562	2.70	6.5	11.6	20.1	22.16	2.87	195.0	171.9	88.2	167.6	160.9	154.3	143.5	57.6	14.6
1.695	2.78	6.7	11.5	19.4	21.11	3.15	191.6	175.5	91.6	169.3	161.8	155.5	146.8	69.9	16.4
0.389	2.73	7.9	14.4	25.0	22.42	3.09	194.9	170.1	87.3	167.5	162.1	154.4	136.6	37.4	10.8
0.641	2.62	7.9	14.7	26.4	21.58	2.96	195.6	171.4	87.6	168.1	162.2	156.2	145.8	68.6	16.5
0.322	2.59	7.9	15.0	26.8	21.37	2.93	196.2	171.4	87.4	167.4	161.2	155.2	144.9	70.8	17.2
0.593	2.67	6.5	11.6	20.3	21.88	2.92	196.6	172.0	87.5	167.2	160.8	153.7	141.3	57.9	13.7
1.388	2.78	6.5	12.6	23.7	22.42	2.99	195.2	175.1	89.7	170.8	165.2	159.4	149.1	58.0	14.7
0.23	2.99	6.2	11.4	20.4	21.27	2.98	196.0	175.5	89.6	171.2	166.3	159.9	137.2	32.4	9.3

Last line: A123 control powder; Other lines: WPI recycled powder

Recovered NMC622 powder has comparable performance comparing to the commercial powder. The high rate performance of recovered powder is better.

Phase II: A123 Test Results (Single Layer Pouch Cells)



Test Request #	WPI Lot #
TXS10222	12172018-C1
TXS10227	12172018-C2
TXS10228	12172018-C3
TXS10229	Control

- Data very consistent between groups
- Observed slight increase in capacity for all groups except for TXS10227 (green)

Scale-up and Commercialization

Battery Resourcers Inc

- Design capacity: 500 kg/day
- 6000 ft^2 in Worcester, MA
- Operational since Summer 2018

Responses to Previous Year Reviewers' Comments

Comment 1: The reviewer would have liked some discussion on how the technology would be commercially viable as electrodes become better and better and cells last longer.

Response: Although the electrodes become better and better and cells last longer, the EV batteries still last ~10 years. In fact, Battery Resourcers has studied the market of spent EV Batteries and found the quantity is pretty significantly.

Comment 2: The reviewer found the approach to be okay, but the target sort of moved over the course of the program. In retrospect, the program should have addressed the robustness of the process to changes in feed stream or changes in the Ni ratio in the resulting precursors. The precursors critical to quality characteristics were not clearly stated and may not have been thoroughly addressed. The reviewer commented that the impact of cell SOH and robustness to commingling cathode chemistries/surface-treated cathodes or even variations from suppliers should have been addressed.

Response: We do not feel that the approach is moved over the course of the program. During the limitation of the poster, we can not present all the data generated in this program. We have generated NMC111 powder from 4 different recycling streams in phase I and each stream could include different batteries with different chemistry. In the phase II program, we are studying how different anode materials and adhesives affect the overall recycling process and recovered NMC powder. All these work is demonstrating how robust the recycling technology is.

Comment 3: The reviewer thought that there needs to be more investigation of cells to avoid hero cell syndrome.

Response: We totally agree that the cells with recycled materials should be investigated very thoroughly. Both A123 and Argonne National Lab have done very detailed testing and characterization of the cells fabricated in this program.

Collaboration and Partners

Scale-up and commercialization

Fabricate commercial cells

Disassemble EV battery packs

Evaluate cells fabricated with recycled materials

Provide battery packs

Remaining Challenges and Barriers

- Understand the long term performance of the recycled NMC622
- Understand the coating performance
- Determine the impacts of different anode materials and adhesives

Proposed Future Work

- Determine the coating strategy based on A123 SLP cycle results
- Scale up recycling experiments to generate powder for 1Ah and 10Ah cell fabrication
- Determine the impacts of different anode and adhesive materials
- Optimize the economical model

Summary

- High performance NMC622 has been recovered from spent lithium ion batteries
- Different coating methods were developed to stabilize NMC622
- The coating of NMC622 is uniform, and does not sacrifice the electrochemical performance from the preliminary results
- WPI NMC622 with/without coating normally shows better rate performance, which is consistent with the phase I results on NMC111